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硕 士 学 位 论 文

厦门市大气汞干湿沉降及汞污染源的探究

Dry and Wet Deposition Characteristics and Anthropogenic
Sources of Atmospheric Mercury in Xiamen

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摘要

大气汞是危害人体健康的毒性污染物。大气中不同形态的汞具有不同的理化特性,颗粒态总汞 TPM(Total particulate mercury)沉降速率高,能在排放源附近沉降,对区域内的生态环境造成较大的危害;大气气态总汞 TGM(Total gaseous mercury)因其具有较低的水溶性和沉降速率,可在大气中停留 0.5-2 年,能够进行长距离传输,被认为是全球性污染物。目前欧美地区已经广泛地建立起汞监测网络,虽然国内的大气汞监测也陆续开展起来,但是长时间持续监测尚不多见。本研究基于实验室前期的研究成果,于 2014 年 8 月至 2016 年 2 月在厦门大学思明校区及翔安校区设点监测 TGM、TPM,同时利用 HYSPLIT 模型及 ArcGIS 软件探讨不同轨迹的大气气团的影响,并基于排放因子法和既有的统计数据建立厦门市汞排放清单,以进一步了解厦门市大气汞的时空变化特征和可能存在的污染源。结果如下:

监测表明,思明校区和翔安校区 TGM 浓度分别 $3.58 \pm 1.30 \text{ ng/m}^3$ 和 $3.14 \pm 1.29 \text{ ng/m}^3$, 低于国内典型的工业及矿业城市,但二者的监测值却是北半球大气汞背景浓度的 1.8-2.4 倍;而两个监测点的 TPM 浓度分别为 $168.50 \pm 90.83 \text{ pg/m}^3$ 和 $211.52 \pm 115.35 \text{ pg/m}^3$, 都超过全球颗粒态汞背景值的 2 倍。

厦门市大气汞浓度具有明显的时间分布特征,整体呈现出温暖月份浓度较低、寒冷月份浓度较高,冬春季浓度高、夏秋季浓度低的趋势。季风的变化是影响大气汞浓度月度及季节变化的主要因素,来自北方的气团中含汞量高,能加剧厦门市的大气汞污染;而来自东南海洋的洁净气团能够稀释大气中的污染物。同时,海陆风也是一个重要的因素,由于发生海陆风时污染物的集聚效应,大气汞浓度明显升高(尤其是 TPM)。此外,各个季节的降水、日照时长及大气扩散条件都可能造成大气汞浓度季节性差异。

对厦门市大气汞浓度与气象参数和大气污染物进行相关性分析及主成分分析,发现大气汞浓度与 SO_2 、 NO_2 、CO、 PM_{10} 和 $\text{PM}_{2.5}$ 之间呈现正相关关系,这表明化石燃料燃烧可能是厦门地区最重要的污染源。主成分分析显示,不同季节受到燃煤排放的影响最大,而大气光化学行为及交通尾气排放可能是潜在的影响因素。

将厦门大气 TGM 监测数据分为高浓度组和低浓度组，分别表示浓度值为最大 15%的数据集和浓度值为最小 15%的数据集，利用 HYSPLIT 后推轨迹模型和 ArcGIS 软件对高浓度组和低浓度组大气气团轨迹进行分类整合。分析显示，高浓度组下，超过 80%的气团轨迹起源于中国内陆地区或气团轨迹经过长三角地区；而低浓度组下，超过 56%的气团轨迹来自海洋。这表明内陆或长三角地区可能是厦门大气 TGM 的主要输入源。

基于排放因子法及相关的统计数据，核算厦门市人为汞排放源。研究表明，2014 年厦门市人为汞排放为 655.59 kg，主要来自于燃煤、垃圾焚烧和节能灯制造，其贡献率分别为 60.67%、19.15%和 17.20%。从 2009 年至 2014 年，厦门市人为汞排放整体呈现上升趋势，汞排放量取决于经济发展情况及厦门市垃圾焚烧电厂的扩建运行。2015 年以来，随着燃煤消耗量的大幅降低，厦门市大气 TGM 和 TPM 浓度比 2014 年分别下降 29.0%和 22.2%。

对大气汞干湿沉降的实地监测有利于进一步理解其沉降特性。2014 年 5 月至 2015 年 11 月期间，S2 点降水中总汞 THg(Total mercury)浓度为 28.14 ± 15.92 ng/L，溶解汞 DHg(Dissolved mercury)浓度为 20.91 ± 14.45 ng/L，全年大气汞湿沉降通量为 $38.44 \mu\text{g}/\text{m}^2$ 。从 2015 年 4 月至 2016 年 2 月，S1 和 S2 点 TPM 沉降速率分别为 1.24 cm/s 和 1.27 cm/s，由此推断厦门市全年大气 TPM 干沉降通量为 $53.14\text{-}66.70 \mu\text{g}/\text{m}^2$ ，由此估算全市 TPM 沉降量为 83.59-104.92 kg。

关键词：厦门；大气汞；时空分布；排放源统计；干湿沉降

Abstract

Atmospheric mercury is a toxic pollutant concerned by human's health. Different speciated mercury behave quite differently, total particulate mercury (TPM) has high deposition velocity, it can deposit around emission sources by wet and dry deposition and cause regional environmental pollution. Total gaseous mercury (TGM) can transport globally and stay in the atmosphere with a residence time of 0.5-2.0 years, which is considered as the most important form of mercury causing serious pollution problem worldwide. Recently, lots of mercury monitoring networks have been established in Europe and North America. Although some field measurements of atmospheric mercury were conducted in China, few of them were long duration. Based on the previous study of our laboratory, this investigation was conducted to monitor TGM and TPM from August 2014 to February 2016, at Siming and Xiangan campus, Xiamen University. In order to further understand the special and temporal distribution and possible sources of atmospheric mercury, HYSPLIT model and ArcGIS software were used to trace air masses, and it also compiled mercury emission inventory with emission factors and official statistical data.

The conclusions are as follows:

During the study period, average TGM concentrations were $3.58 \pm 1.30 \text{ ng/m}^3$ and $3.14 \pm 1.29 \text{ ng/m}^3$ in S1 and S2, respectively. Although the TGM concentration level was lower than those in some typical industrial and mining cities in China, it was approximately 1.8-2.4 times of that in the North Hemisphere background regions. Moreover, the average TPM concentrations were $168.50 \pm 90.83 \text{ pg/m}^3$ and $211.52 \pm 115.35 \text{ pg/m}^3$ in S1 and S2 respectively, which were more than 2 times of that observed in the global background.

The atmospheric mercury showed significant monthly and seasonal distribution patterns, with lower concentrations observed in warm months and higher in cold months, lower in summer and autumn and high in winter and spring. The monsoon weather played a critical role in monthly and seasonal trends. When major prevailing winds were blown from the northeast, atmospheric mercury concentration increased

sharply due to the air masses contained high content mercury. While, the air pollutants could be better dispersed by the clear air masses from the southeastern open ocean. Furthermore, sea-land breeze was the other important factor, the concentration of atmospheric mercury(especially TPM) was easily gathered and increased during the conversion between sea and land breeze. The results also showed that precipitation, sunshine duration and dispersion condition contributed to variation of atmospheric mercury concentration in different seasons.

This study conducted the correlation analysis and principal component analysis(PCA) between atmospheric mercury and meteorological parameters and/or criteria air pollutant concentrations. The concentration of atmospheric mercury correlated positively with SO₂, NO₂, CO, PM₁₀ and PM_{2.5}, which indicated that fossil fuel combustion was the most important anthropogenic source of atmospheric mercury in Xiamen. Furthermore, the PCA result indicated the coal combustion contributed mostly to TGM and TPM pollution in each season, and photochemical processing and vehicle exhaust were potential sources.

The HYSPLIT backward trajectory model and ArcGIS software were conducted to trace transportation routes of air masses at Xiamen. TGM data were clustered into two categories: (1) high concentration set with values ranked top 15% of all data, and (2) low concentration set with values ranked last 15%. And the analysis demonstrated that more than 80% of the air masses trajectories originated from inland China or went through Yangtze River Delta(YRD) region when high concentration set occurred. However, more than 56% of air masses come from open ocean when low concentration set occurred. It suggested that North China and YRD region may be the important mercury source areas for Xiamen.

The mercury emission statistics in Xiamen was estimated by using emission factors and official statistical data. It suggested that mercury emission from anthropogenic sources was up to 655.59 kg for 2014, and the coal combustion, solid-waste incineration and lamp product accounted for 60.67%, 19.15% and 17.20% of the total. The mercury emitted from anthropogenic sources increased gradually from 2009 to 2014, and the emission was depended mainly on the economical

condition and the new waste incinerators. However, due to the decreasing coal consumption, the atmospheric TGM and TPM concentrations fell 29.0% and 22.2% in 2015 compared to those in 2014, respectively.

Field measurement of speciated mercury in precipitation and dry deposition of TPM were conducted to better understand the characteristics of mercury deposition. This study showed that total mercury (THg) concentration was 28.14 ± 15.92 ng/L, and the dissolved mercury (DHg) concentration was 20.91 ± 14.45 ng/L in precipitation, from May 2014 to November 2015. Wet deposition of mercury in S2 was estimated to be $38.44 \mu\text{g}/\text{m}^2$. From April 2014 to February 2015, the TPM dry deposition velocities were 1.24 cm/s and 1.27 cm/s, and it was believed that the range of dry deposition flux from 53.14 to $66.70 \mu\text{g}/(\text{m}^2 \cdot \text{a})$ in Xiamen.

Key words: Xiamen; Atmospheric mercury; Temporal and special distribution; Emission statistics; wet and dry deposition.

第1章 绪论

汞(Mercury, Hg), 俗称水银, 是最早被人认识及应用的金属元素之一。汞的用途广泛, 先秦时期我国先民开始使用水银对铜器进行鎏金装饰, 随着儒家及道教的兴起和发展, 丹砂(HgS)逐渐被应用于绘画和炼丹。工业革命以来, 汞作为农药、金属电解电极、电器及仪表制造的必要金属, 在工农业生产领域中发挥着不可替代的作用。但是汞又是一种致畸、致癌、致基因突变的元素, 对人类及生态环境造成严重的危害, 特别是 20 世纪 50 年代日本水俣病(Minamata disease)的爆发, 汞的环境问题逐渐受到关注。

1.1 汞的理化性质及生物毒性

1.1.1 汞的物理化学行为

汞的元素序数 80, 处于元素周期表中的第 6 周期、第 II_B 族, 它的熔点及沸点分别是 -38.9 °C 和 356.9 °C, 相对原子质量为 200.59。在常温常压下汞是唯一一种呈现液态的金属元素, 并具有银白色光泽。

在大气环境中, 汞主要以 Hg^0 、 Hg^+ 和 Hg^{2+} 三种价态形式存在^[1]。由于汞的电子构型是 $[\text{Xe}]4f^{14}5d^{10}6s^2$, 容易失去最外层轨道 $6s^2$ 中的电子, 形成 +2 价态, 所以在自然环境中, 汞主要以元素态 Hg^0 和氧化态 Hg^{2+} 的价态存在, 而处于中间价态的 Hg^+ 极不稳定。 Hg^0 具有较高的挥发性和较强烈的脂溶性; Hg^+ 多以二聚体(Hg_2^{2+})的形式存在; Hg^{2+} 的化合物包括 HgO 、 HgS 、 HgCl_2 、 HgI_2 、 $\text{Hg}(\text{NO}_3)_2$ 、 HgSO_4 等。汞能够溶解多种金属(如钾、钠、金、银等), 形成类似于合金的形态, 称为汞齐^[2], 常见的汞齐有钠汞齐、锌汞齐、铜汞齐、银锡汞齐、金汞齐等。

1.1.2 汞的生物毒性

20 世纪 50 年代日本发生了大规模汞中毒事件, 绝大多数患者是通过饮食(尤其是食用含有甲基汞的鱼类、贝类等的渔业产品)摄入过量的汞, 引起汞在肝、肾的蓄积, 导致脏器功能的衰竭, 还可能累积在脑组织中, 造成永久性中枢神经损伤。

不同形态的汞对人体毒性并不相同, 元素汞毒性较小, 无机汞次之, 有机汞最强, 其中甲基汞的致病性最为严重。甲基汞中毒的化学生物理论认为, 进入人体的甲基汞能与蛋白质和反应酶的巯基相结合, 形成硫醇盐, 从而抑制含 -SH 基

的酶的活性^[3],破坏细胞的基本功能和代谢。甲基汞的强毒性是由它的性质决定的:(a)甲基汞具有亲脂性,易溶于脂肪和类脂质中。研究显示,甲基汞在肠道中的吸收率达到100%,能够溶解在血液红细胞膜脂类及血红蛋白中,随血液输送至全身^[4];(b)甲基汞的C-Hg键很牢固,在体内不易断键,在细胞中能够保持原形,医学上称之为原形蓄积^[4];(c)甲基汞具有强神经毒性,能够促使游离的 Ca^{2+} 进入神经细胞胞浆中,诱导神经细胞凋亡^[5]。

汞对于植物的生长发育也存在一定的影响^[6]。高浓度的汞($5\text{ }\mu\text{g/mL}$ 及以上)不仅会抑制种子的萌发,降低根部和茎部的长度和质量,使植物生长减缓,也会增加植物体内的光合作用色素粒和叶绿素含量,影响光合作用中电子传递,抑制植物的光合作用。

1.2 大气汞的形态及迁移转化

1.2.1 大气汞形态分类

在大气环境中,根据汞的物理化学形态的差异可以分为:气态元素汞(Hg^0 或GEM, elemental gaseous mercury)、活性气态汞(Hg^{2+} 或者RGM, reactive gaseous mercury)和颗粒态汞($\text{Hg}(\text{p})$ 或TPM, total particulate mercury)^[7-10],其中气态元素汞 Hg^0 和活性气态汞 Hg^{2+} 统称为气态总汞(TGM, total gaseous mercury),气态总汞TGM和颗粒态汞TPM统称为总大气汞(TAM, total atmospheric mercury)。

Hg^0 是大气中最主要的汞形态,约占大气TAM的90%以上^[1, 11]。 Hg^0 水溶性极低,不会因湿沉降而被去除,在对流层中的停留时间长达0.5-2.0年^[8, 10]。因此, Hg^0 可经由大气长程传输参与全球汞循环,是一种被公认的全球性大气污染物。

Hg^{2+} 是由气相或液相中的 Hg^0 被 O_3 、 $\cdot\text{OH}$ 等氧化剂氧化形成的,在实验操作上被定义为能吸附在镀KCl扩散管上的汞^[9, 10]。虽然 Hg^{2+} 只占TGM的1-3%^[8, 11],但其水溶性比 Hg^0 高3个数量级,因此 Hg^{2+} 很容易随着降水而去除,扩散范围通常在排放源周围几十到几百公里之内,是大气汞干湿沉降的主要贡献者。 Hg^{2+} 浓度是大气汞干湿沉降速率的重要影响因子,同时也是评估区域内大气汞对生态影响的主要因子^[12]。

TPM是吸附在大气颗粒物及海洋气溶胶上的元素汞 Hg^0 、活性气态汞 Hg^{2+} 和/或有机汞等^[13, 14]。尽管TPM在TAM中的比重小于10%^[1, 11],但由于其较高

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